

Quantum optical non-linearities induced by Rydberg-Rydberg interactions : a perturbative approach

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In this article, we theoretically study the quantum statistical properties of the light transmitted through or reflected from an optical cavity, filled by an atomic medium with strong optical non-linearity induced by Rydberg-Rydberg van der Waals interactions. Atoms are driven on a two-photon transition from their ground state to a Rydberg level via an intermediate state by the combination of a weak signal field and a strong control beam. By using a perturbative approach, we get analytic results which remain valid in the regime of weak feeding fields, even when the intermediate state becomes resonant. Therefore they allow us to investigate quantitatively new features associated with the resonant behaviour of the system. We also propose an effective non-linear three-boson model of the system which, in addition to leading to the same analytic results as the original problem, sheds light on the physical processes at work in the system.

I. INTRODUCTION

Photons are considered as the best long-range quantum information carriers; they, however, do not directly interact with each other, which makes the processing of the information they carry problematic [1]. Standard Kerr dispersive non linearities obtained in non-interacting atomic ensembles, either in off-resonant two-level or resonant three-level configurations involving Electromagnetically Induced Transparency (EIT), are usually too small to allow for quantum non-linear optical manipulations. Among other techniques [1], a possible way to enhance the non-linear susceptibility is to resort to a Rydberg level as one of the long-lived states involved in the EIT process [2–6] : in such Rydberg EIT protocols, the strong van der Waals interactions between Rydberg atoms create a cooperative Rydberg blockade phenomenon [7–9], where each Rydberg atom prevents the excitation of its neighbors inside a "blockade sphere" and deeply changes the EIT profile. In particular, giant dispersive non-linear effects were experimentally obtained in an off-resonant Rydberg-EIT scheme using cold rubidium atoms placed in an optical cavity [10, 11]. In a previous paper [6], we theoretically investigated the quantum statistical properties of the light generated by this scheme in the dispersive regime, *i.e.* for strongly detuned intermediate state. We showed that, under some assumptions, the system effectively behaves as a large spin coupled to the cavity mode [12] and we computed the steady-state second-order correlation function to characterize the bunched or antibunched emission of photons out of the cavity.

In the present paper, we deal with the same system, but in a different approach. Restricting ourselves to the low feeding regime, we present an analytic derivation of the correlation function $g^{(2)}(\tau)$ for the transmitted and reflected light, based on the factorization of the lowest perturbative order of operator product averages. It is important to note that this derivation is valid in both the dispersive and resonant regimes and therefore gener-

alizes our previous results. This factorization property is demonstrated rigorously for purely radiative damping, but we show also that it is approximately preserved in the experimentally relevant case of additional dephasing due to, e.g., laser frequency and intensity noise. In addition, we propose an effective non-linear three-boson model for the coupled atom-cavity system which allows us to obtain the same results as the (more cumbersome) exhaustive treatment. In the dispersive regime, this Hamiltonian agrees with the one we obtained in the so-called "Rydberg-bubble approximation" [6]; it also allows us to investigate the dissipation at work in the resonant case.

The paper is structured as follows. In Sec. II, we recall our setup and the assumptions we make to compute its dynamics. In Sec. III, we present an analytical way to obtain the correlation functions for the light outgoing from the cavity and discuss some of the numerical results we obtained. In Sec. IV, we present and discuss an effective three-boson model, allowing us to recover and generalize the previous results. Finally, we conclude in Sec. V by evoking open questions and perspectives of our work. Appendices address supplementary technical details which are omitted in the text for readability.

II. THE SYSTEM

The system we consider here is the same we dealt with in [6]. It comprises N atoms which present a three-level ladder structure with a ground $|g\rangle$, intermediate $|e\rangle$ and Rydberg states $|r\rangle$ (see Figure 1). The energy of the atomic level $|k = g, e, r\rangle$ is denoted by $\hbar\omega_k$ (by convention $\omega_g = 0$) and the dipole decay rates are γ_e (intermediate state) and γ_r (Rydberg state). The transitions $|g\rangle \leftrightarrow |e\rangle$ and $|e\rangle \leftrightarrow |r\rangle$ are respectively driven by a weak probe field of frequency ω_p and a strong control field of frequency ω_{cf} . Both fields can *a priori* be resonant or not with atomic transitions, the respective detunings being defined by $\Delta_e \equiv (\omega_p - \omega_e)$ and

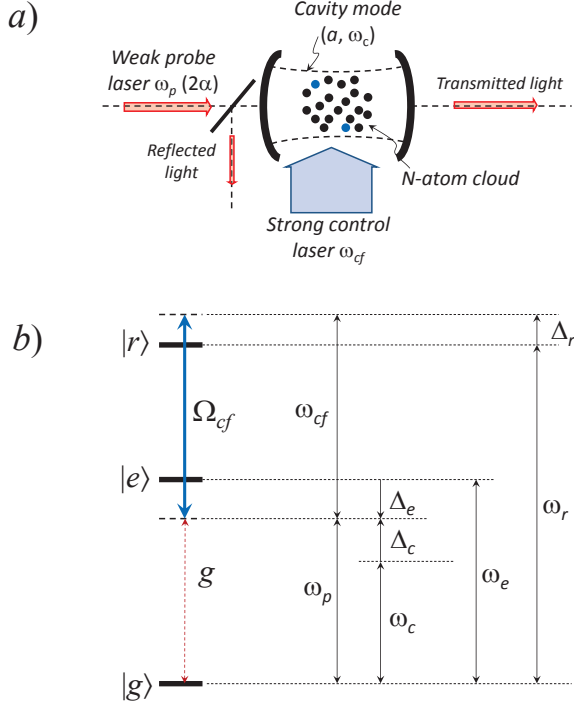


FIG. 1. a) The setup consists of N cold atoms placed in an optical cavity which is fed by a weak (classical) laser beam of frequency ω_p and a strong control laser field of frequency ω_{cf} . b) The atoms present a three-level ladder structure $\{|g\rangle, |e\rangle, |r\rangle\}$. The transitions $|g\rangle \leftrightarrow |e\rangle$ and $|e\rangle \leftrightarrow |r\rangle$ are driven by the injected probe and control laser fields, respectively, with the respective coupling strength and Rabi frequency g and Ω_{cf} (see the text for the definitions of the different detunings represented here).

$\Delta_r \equiv (\omega_p + \omega_{cf} - \omega_r)$. Moreover, the atoms are placed in an optical cavity: we shall denote by $\gamma_c^{(L,R)}$ the respective decay rates through the left and right mirrors (see Fig. 1), with $\gamma_c \equiv \gamma_c^{(L)} + \gamma_c^{(R)}$. The transition $|g\rangle \leftrightarrow |e\rangle$ is supposed in the neighborhood of a cavity resonance. The frequency and annihilation operator of the corresponding mode are denoted by ω_c and a , respectively; the detuning of this mode with the probe laser is defined by $\Delta_c \equiv (\omega_p - \omega_c)$ and α denotes the feeding rate of the cavity mode with the probe field, which is supposed real for simplicity. Finally, we introduce g and Ω_{cf} which are the single-atom coupling constant of the transition $|g\rangle \leftrightarrow |e\rangle$ with the cavity mode and the Rabi frequency of the control field on the transition $|e\rangle \leftrightarrow |r\rangle$, respectively. As represented on Figure 1, the setup allows one to measure the statistics of both the reflected and transmitted lights, i.e. $g^{(2)}(\tau)$.

The dynamics of the full system, including the bath modes, are governed by the Hamiltonian derived in Appendix A, in the Rotating Wave Approximation. We note that this Hamiltonian description does not take into account any additional dephasing due to, e.g., laser intensity or frequency fluctuations: decays and dephasing are therefore purely radiative. In the Markov approximation, the corresponding Heisenberg-Langevin equations are

$$\frac{d}{dt} a = iD_c a - i\alpha - ig \sum_i \sigma_{ge}^{(i)} + \sqrt{2\gamma_c^{(L)}} a_{in}^{(L)} + \sqrt{2\gamma_c^{(R)}} a_{in}^{(R)} \quad (1)$$

$$\frac{d}{dt} \sigma_{ge}^{(i)} = iD_e \sigma_{ge}^{(i)} - i\frac{\Omega_{cf}}{2} \sigma_{gr}^{(i)} + ig a (\sigma_{ee}^{(i)} - \sigma_{gg}^{(i)}) + F_{ge}^{(i)} \quad (2)$$

$$\frac{d}{dt} \sigma_{gr}^{(i)} = iD_r \sigma_{gr}^{(i)} - i\frac{\Omega_{cf}}{2} \sigma_{ge}^{(i)} + ig a \sigma_{er}^{(i)} - i\sigma_{gr}^{(i)} \sum_{j \neq i}^N \kappa_{ij} \sigma_{rr}^{(j)} + F_{gr}^{(i)} \quad (3)$$

$$\frac{d}{dt} \sigma_{er}^{(i)} = iD_{er} \sigma_{er}^{(i)} + i\frac{\Omega_{cf}}{2} (\sigma_{rr}^{(i)} - \sigma_{ee}^{(i)}) + ig a^\dagger \sigma_{gr}^{(i)} - i\sigma_{er}^{(i)} \sum_{j \neq i}^N \kappa_{ij} \sigma_{rr}^{(j)} + F_{er}^{(i)} \quad (4)$$

where $a_{in}^{(L)}$, $a_{in}^{(R)}$ and $F_{\alpha\beta}^{(i)}$ denote Langevin forces associated to the incoming fields from left and right sides and to the atomic operator $\sigma_{\alpha\beta}^{(i)}$, respectively. We also introduced the complex effective detunings $D_k \equiv (\Delta_k + i\gamma_k)$ for $k = c, e, r$ and $D_{er} \equiv (\Delta_r - \Delta_e) + i(\gamma_r + \gamma_e)$. Note that we chose to make the feeding factor α appear explicitly in Eq.(1): in technical terms, it corresponds to displacing the incoming field from the coherent state $|\alpha\rangle$ to the vacuum $|0\rangle$; to be consistent with this choice, from now on, we must set $\langle a_{in} \rangle = 0$.

In the next section, we show how to compute the correlation function $g^{(2)}(\tau)$ at the lowest order in the feeding

parameter α for the transmitted and reflected light.

III. PERTURBATIVE CALCULATION OF $g^{(2)}$

A. Correlation functions of the transmitted and reflected light.

The second-order correlation function characterizes the bunched ($g^{(2)}(0) > (g^{(2)}(\tau))$) or anti-bunched ($g^{(2)}(0) < (g^{(2)}(\tau))$) nature of the light transmitted or reflected by the cavity. For the transmitted light on the right side (R) of the cavity, one has by definition $g_t^{(2)}(0) \equiv \langle a_{out}^{(R)\dagger} a_{out}^{(R)\dagger} a_{out}^{(R)} a_{out}^{(R)} \rangle / \langle a_{out}^{(R)\dagger} a_{out}^{(R)} \rangle^2$, where $a_{out}^{(R)}$ is the transmitted mode field annihilation operator, and all averages should be evaluated in the steady state. From the input-output relations [13], one gets $a_{out}^{(R)} + a_{in}^{(R)} = \sqrt{2\gamma_c^{(R)}} a$, and hence

$$g_t^{(2)}(0) = \langle a^\dagger a^\dagger a a \rangle / \langle a^\dagger a \rangle^2.$$

For the reflected light on the left side (L) of the cavity, one gets $g_r^{(2)}(0) \equiv \langle a_{out}^{(L)\dagger} a_{out}^{(L)\dagger} a_{out}^{(L)} a_{out}^{(L)} \rangle / \langle a_{out}^{(L)\dagger} a_{out}^{(L)} \rangle^2$, where $a_{out}^{(L)}$ is the reflected mode field annihilation operator. Similarly, by using the input-output relation

$$a_{out}^{(L)} + a_{in}^{(L)} - i \frac{\alpha}{\sqrt{2\gamma_c^{(L)}}} = \sqrt{2\gamma_c^{(L)}} a$$

for the left mirror, one gets

$$\begin{aligned} \langle a_{out}^{(L)\dagger} a_{out}^{(L)\dagger} a_{out}^{(L)} a_{out}^{(L)} \rangle &= (2\gamma_c^{(L)})^2 \langle a^\dagger a^\dagger a a \rangle + \\ &4i\alpha\gamma_c^{(L)} [\langle a^\dagger a^\dagger a \rangle - \langle a^\dagger a a \rangle] + i \frac{\alpha^3}{\gamma_c^{(L)}} (\langle a^\dagger \rangle - \langle a \rangle) + \\ &\alpha^2 (4 \langle a^\dagger a \rangle - \langle a^\dagger a^\dagger \rangle + \langle a a \rangle) + \frac{\alpha^4}{(2\gamma_c^{(L)})^2} \\ \langle a_{out}^{(L)\dagger} a_{out}^{(L)} \rangle &= 2\gamma_c^{(L)} \langle a^\dagger a \rangle + i\alpha (\langle a^\dagger \rangle - \langle a \rangle) + \frac{\alpha^2}{2\gamma_c^{(L)}} \end{aligned}$$

B. Factorization in the perturbative limit

In the whole paper, we shall restrict ourselves to the low excitation regime, *i.e.* to low values of the feeding parameter α . We therefore seek $g^{(2)}(0)$ at the lowest non-vanishing order in α : this requires to evaluate $\langle a^\dagger a^\dagger a a \rangle$, $\langle a^\dagger a^\dagger a \rangle$ and $\langle a^\dagger a \rangle$ at the fourth, third and second orders, respectively. This task is greatly simplified by the following remarkable factorization property, established in Appendix B,

$$\begin{aligned} \langle a^\dagger(t) a(t) \rangle^{(2)} &= \langle a^\dagger(t) \rangle^{(1)} \langle a(t) \rangle^{(1)} \\ \langle a^\dagger(t_2) a^\dagger(t_1) a(t_1) \rangle^{(3)} &= \langle a^\dagger(t_2) a^\dagger(t_1) \rangle^{(2)} \times \langle a(t_1) \rangle^{(1)} \\ \langle a^\dagger(t_2) a^\dagger(t_1) a(t_1) a(t_2) \rangle^{(4)} &= \\ &\langle a^\dagger(t_2) a^\dagger(t_1) \rangle^{(2)} \times \langle a(t_1) a(t_2) \rangle^{(2)} \end{aligned}$$

where the superscript (k) denotes the order in α to which quantities are calculated. Therefore, for instance, for the transmitted light,

$$g_t^{(2)}(0) = \left(\langle a^\dagger a^\dagger \rangle^{(2)} \langle a a \rangle^{(2)} \right) / \left(\langle a^\dagger \rangle^{(1)} \langle a \rangle^{(1)} \right)^2$$

and we merely need to determine $\langle a \rangle^{(1)}$ and $\langle a^2 \rangle^{(2)}$. Note that the factorization does not apply to products of the kind $\langle a^2 \rangle^{(2)}$, so that $\langle a^2 \rangle^{(2)} \neq \langle a \rangle^{(1)} \langle a \rangle^{(1)}$.

The mean values $\langle a \rangle^{(1)}$ and $\langle \sigma_{ge}^{(i)} \rangle^{(1)}$ are readily obtained through taking the steady state of the first-order averaged Heisenberg equations Eqs. (1-4)

$$\langle a \rangle^{(1)} = \frac{\alpha}{D_c - \frac{g^2 N}{\left(D_e - \frac{\Omega_{cf}^2}{4D_r}\right)}} \quad (5)$$

$$\langle \sigma_{ge}^{(i)} \rangle^{(1)} = \frac{\alpha g}{D_c \left(D_e - \frac{\Omega_{cf}^2}{4D_r}\right) - g^2 N} \quad (6)$$

$$\langle \sigma_{gr}^{(i)} \rangle^{(1)} = \frac{\alpha g \Omega_{cf}}{2D_r \left[D_c \left(D_e - \frac{\Omega_{cf}^2}{4D_r}\right) - g^2 N\right]} \quad (7)$$

The second-order value $\langle a^2 \rangle^{(2)}$ is determined through solving the following closed system

$$\langle a^2 \rangle^{(2)} = \frac{g\sqrt{N}}{D_c} \langle ab \rangle^{(2)} + \frac{\alpha}{D_c} \langle a \rangle^{(1)} \quad (8)$$

$$\langle ab \rangle^{(2)} = \frac{\Omega_{cf}}{2(D_c + D_e)} \langle ac \rangle^{(2)} + \frac{g\sqrt{N}}{(D_c + D_e)} \langle aa \rangle^{(2)} + \frac{g\sqrt{N}}{(D_c + D_e)} \langle bb \rangle^{(2)} + \frac{\alpha}{(D_c + D_e)} \langle b \rangle^{(1)} \quad (9)$$

$$\langle ac \rangle^{(2)} = \frac{g\sqrt{N}}{(D_c + D_r)} \langle bc \rangle^{(2)} + \frac{\alpha}{(D_c + D_r)} \langle c \rangle^{(1)} + \frac{\Omega_{cf}}{2(D_c + D_r)} \langle ab \rangle^{(2)} \quad (10)$$

$$\langle bb \rangle^{(2)} = \frac{\Omega_{cf}}{2D_e} \langle bc \rangle^{(2)} + \frac{g\sqrt{N}}{D_e} \langle ab \rangle^{(2)} \quad (11)$$

$$\langle bc \rangle^{(2)} = \frac{\Omega_{cf}}{2(D_e + D_r)} \langle cc \rangle^{(2)} + \frac{g\sqrt{N}}{(D_e + D_r)} \langle ac \rangle^{(2)} + \frac{\Omega_{cf}}{2(D_e + D_r)} \langle bb \rangle^{(2)} \quad (12)$$

$$\langle cc \rangle^{(2)} = \frac{\Omega_{cf}g\sqrt{N}}{2} K \langle ac \rangle^{(2)} + \frac{\Omega_{cf}^2g\sqrt{N}}{4D_e} K \langle ab \rangle^{(2)} \quad (13)$$

deduced from Eqs. (1-4) under the assumption of an homogeneous atomic medium, whose consequences are detailed in Appendix C. In this system, we introduced the collective atomic operators

$$b \equiv \frac{1}{\sqrt{N}} \sum_i \sigma_{ge}^{(i)} \quad c \equiv \frac{1}{\sqrt{N}} \sum_i \sigma_{gr}^{(i)}.$$

We note that the first-order mean values $\langle a \rangle^{(1)}$, $\langle b \rangle^{(1)}$ and $\langle c \rangle^{(1)}$ which appear in Eqs. (8, 9, 10), respectively, have been computed in Eqs. (5, 6, 7). The K coefficient is approximately given by (see Appendix C for details)

$$K \approx \frac{1}{\left(D_e + D_r - \frac{\Omega_{cf}^2}{4D_e}\right) D_r - \frac{\Omega_b^2}{4}} \left(1 - \frac{V_b}{V}\right) \quad (14)$$

where

$$V_b = \frac{\sqrt{2}\pi^2}{3} \sqrt{\frac{-C_6}{D_r - \Omega_{cf}^2 / \left(4(D_e + D_r) - \frac{\Omega_{cf}^2}{D_e}\right)}} \quad (15)$$

will be interpreted as the Rydberg bubble volume in the dispersive regime in the next section. Though it is too cumbersome to be reproduced here, the solution for $\langle a^2 \rangle^{(2)}$ is simply obtained by matrix inversion, and the calculation of $g_t^{(2)}(0)$ and $g_r^{(2)}(0)$ can be straightforwardly programmed, e.g. in Mathematica.

As it has been the case for $g_{t,r}^{(2)}(0)$, the calculation of the time-dependent correlation function $g_{t,r}^{(2)}(\tau) \equiv \langle a^\dagger(t) a^\dagger(t+\tau) a(t+\tau) a(t) \rangle / \langle a^\dagger a \rangle^2$ is greatly simplified by the factorization property derived in Appendix B, since we simply need to determine the quantity $\langle a(t+\tau) a(t) \rangle$. From Eqs. (1-4), one easily deduces the following differential system, at the lowest order in α ,

$$\begin{aligned} \frac{d}{d\tau} \begin{pmatrix} \langle a(t+\tau) a(t) \rangle \\ \langle b(t+\tau) a(t) \rangle \\ \langle c(t+\tau) a(t) \rangle \end{pmatrix} &= -i\alpha \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} + \\ -i \begin{pmatrix} -D_c & g\sqrt{N} & 0 \\ g\sqrt{N} & -D_e & \frac{\Omega_{cf}}{2} \\ 0 & \frac{\Omega_{cf}}{2} & -D_r \end{pmatrix} \begin{pmatrix} \langle a(t+\tau) a(t) \rangle \\ \langle b(t+\tau) a(t) \rangle \\ \langle c(t+\tau) a(t) \rangle \end{pmatrix} \end{aligned}$$

which, together with the initial condition

$$\begin{pmatrix} \langle a(t+\tau) a(t) \rangle \\ \langle b(t+\tau) a(t) \rangle \\ \langle c(t+\tau) a(t) \rangle \end{pmatrix}_{\tau=0} = \begin{pmatrix} \langle aa \rangle^{(2)} \\ \langle ba \rangle^{(2)} \\ \langle ca \rangle^{(2)} \end{pmatrix}$$

calculated above, allows us to determine $\langle a(t+\tau) a(t) \rangle$. Again, though involved, the expressions are straightforward to obtain and program.

C. Application to an experimental case.

1. Dispersive regime.

Let us now provide some numerical results obtained in the perturbative approach described above. We first investigate the dispersive non-resonant regime, addressed in our previous work [6]. To be specific, we consider the same system, namely an ensemble of ^{87}Rb atoms, whose state space is restricted to the levels $|g\rangle = |5s_{\frac{1}{2}}; F=2\rangle$, $|e\rangle = |5p_{\frac{3}{2}}; F=3\rangle$ and $|r\rangle = |95d_{\frac{5}{2}}; F=4\rangle$. The respective radiative decay rates are $\gamma_e = 2\pi \times 3$ MHz and $\gamma_r = 2\pi \times 0.03$ MHz, the cavity decay rate is $\gamma_c = 2\pi \times 1$ MHz, the volume of the sample is $V = 40\pi \times 15^2 \mu\text{m}^3$, the sample density $n_{at} = 0.4 \mu\text{m}^{-3}$, and the cooperativity $C = g^2N/(2\gamma_e\gamma_c) = 1000$.

The other parameters take the same values as in [6]: in units of γ_e , the control laser Rabi frequency is $\Omega_{cf} = 10$, the detuning of the intermediate level is $\Delta_e = -35$, the detuning of the Rydberg level is $\Delta_r = 0.4$, the cavity feeding rate is $\alpha = 0.01$, and the Van der Waals coefficient is $C_6 = -8.83 \times 10^6 \gamma_e \mu\text{m}^6$. For these parameters,

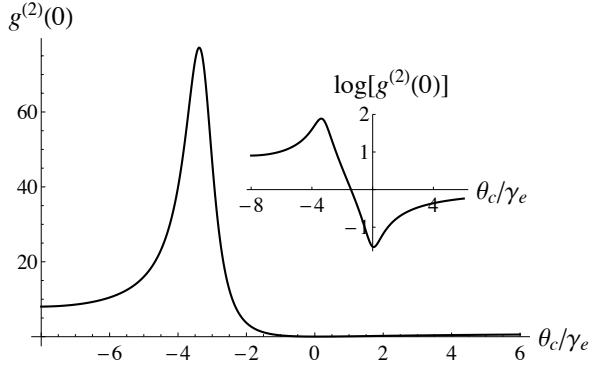


FIG. 2. Second-order correlation function $g_t^{(2)}(0)$ for the transmitted light in the dispersive regime considered in [6] as a function of the renormalized cavity detuning $\theta_c/\gamma_e \equiv (\Delta_c - \Delta_c^{(0)})/\gamma_e$ where $\Delta_c^{(0)}$ is the detuning of the linear cavity. The shape of the plot is in good qualitative agreement with the results of the previous model. Inset : the same plot in logarithmic scale (bunching and antibunching peaks are more clearly visible).

the maximal average number of photons in the cavity is obtained for the cavity detuning $\Delta_c^{(0)} = -6.15206 \gamma_e$ which is taken as a reference.

Let us note however that in real experimental conditions, the atoms undergo not only radiative damping, but are also subject to extra dephasing γ_d on the Rydberg-ground state transition, due to laser frequency and intensity noise. This additional dephasing cannot be modeled in the Hamiltonian formalism presented in Appendix A, and thus the demonstration given in Appendix B for the factorization of mean values does not apply any more. However, since the radiative coherence damping is $\gamma_r \approx 0.01 \gamma_e$, the additional damping is $\gamma_d \approx 0.15 \gamma_e$, and the total number of atoms in the sample is $N \approx 10^4$, the experimental parameters satisfy the condition $\gamma_r \ll \gamma_d \ll N\gamma_r$. Under these circumstances, it is shown in Appendix D that the factorization remains valid, provided that the coherence radiative damping γ_r is replaced by the dephasing rate γ_d in the equations.

Under these conditions, Figure 2 shows the second-order correlation function $g_t^{(2)}(0)$ as a function of the reduced cavity detuning $\theta_c \equiv (\Delta_c - \Delta_c^{(0)})/\gamma_e$, to be compared with Fig. 2 a) in [6]. The two plots are in good qualitative agreement, but the position of the bunching peak is shifted from $\theta_c \approx -5$ to $\theta_c \approx -3.5$, for the same parameters. This basically originates from the definition of V_b in [6], differing from the present one by a factor $\sqrt{2}$.

2. Resonant case

After checking that the present work confirms our previous results, obtained in the dispersive regime, let us consider the resonant case, which could not be treated

before. As a new set of parameters, we take $\Delta_c = \Delta_e = \Delta_r = 0$, and we assume that $\gamma_c^{(R)} \ll \gamma_c^{(L)}$. We also choose a higher principal number $n = 100$ for the Rydberg level, for which $\gamma_r = 0.1\gamma_e$. In addition, we fix $\gamma_c = 0.3\gamma_e$, $C = \frac{g^2 N}{2\gamma_e \gamma_c} \approx 30$ and $V = 50\pi \times 20 \times 20 \mu\text{m}^3$. In this regime, $V_b \approx \frac{\sqrt{2}\pi^2}{3} \sqrt{\frac{-C_6}{D_e}}$ is enhanced, therefore magnified non-linear effects are expected.

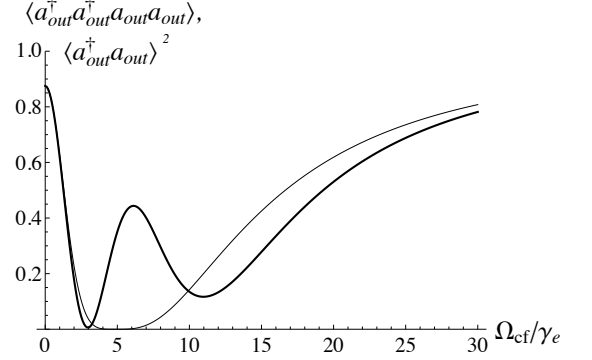


FIG. 3. Resonant case $\Delta_c = \Delta_e = \Delta_r = 0$. The quantities $\langle a_{out}^{(L)\dagger} a_{out}^{(L)} a_{out}^{(L)} a_{out}^{(L)} \rangle$ (thick line) and $\langle a_{out}^{(L)\dagger} a_{out}^{(L)} \rangle$ (thin line), renormalized by the intensity of the incoming light, are represented as functions of the normalized control field Rabi frequency Ω_{cf}/γ_e . For $\Omega_{cf} = 2\sqrt{\gamma_e \gamma_r (2C - 1)} \approx 5\gamma_e$, photon pairs are reflected, i.e. $\langle a_{out}^{(L)\dagger} a_{out}^{(L)} a_{out}^{(L)} a_{out}^{(L)} \rangle \neq 0$, while single photons are absorbed, i.e. $\langle a_{out}^{(L)\dagger} a_{out}^{(L)} \rangle \approx 0$.

As can be seen on Figure 3, there exists a value for which single photons are mostly absorbed $\langle a_{out}^{(L)\dagger} a_{out}^{(L)} \rangle = 0$, while pairs are reflected $\langle a_{out}^{(L)\dagger} a_{out}^{(L)} a_{out}^{(L)} a_{out}^{(L)} \rangle \neq 0$: this value can be computed and is found to be

$$\Omega_{cf} = 2\sqrt{\gamma_e \gamma_r (2C - 1)} = 2\gamma_e \sqrt{6} \approx 5\gamma_e$$

On the contrary, in a slightly detuned case, i.e. for $\Delta_e = -2\gamma_e$ and $\Delta_r = -0.1\gamma_e$, the other parameters remaining the same, one observes that around $\Omega_{cf} \approx 11\gamma_e$ pairs are absorbed $\langle a_{out}^{(L)\dagger} a_{out}^{(L)} a_{out}^{(L)} a_{out}^{(L)} \rangle = 0$ while single photons are reflected $\langle a_{out}^{(L)\dagger} a_{out}^{(L)} \rangle \neq 0$ (see Fig. 4).

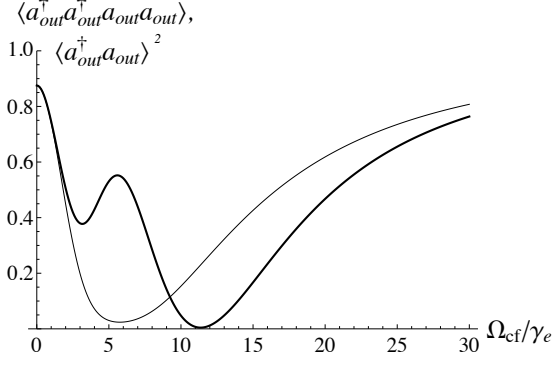


FIG. 4. *Slightly detuned case* $\Delta_c = 0$, $\Delta_e = -2\gamma_e$, $\Delta_r = -0.1\gamma_e$. The quantities $\langle a_{out}^{(L)\dagger} a_{out}^{(L)\dagger} a_{out}^{(L)} a_{out}^{(L)} \rangle$ (thick line) and $\langle a_{out}^{(L)\dagger} a_{out}^{(L)} \rangle$ (thin line), renormalized by the intensity of the incoming light, are represented as functions of the normalized control field Rabi frequency Ω_{cf}/γ_e . For $\Omega_{cf} \approx 11\gamma_e$, photon pairs are absorbed, *i.e.* $\langle a_{out}^{(L)\dagger} a_{out}^{(L)\dagger} a_{out}^{(L)} a_{out}^{(L)} \rangle = 0$, while single photons are reflected, *i.e.* $\langle a_{out}^{(L)\dagger} a_{out}^{(L)} \rangle \neq 0$.

These new features are specific of the near-resonant regime, and were not present in our previous work. They may be interpreted as different impedance matching conditions for single photons and for pairs, leading to very large non-linear losses, acting at the single photon level.

To conclude this section, we described how to obtain the exact and analytic expression of the correlation function in the low excitation regime, valid not only in the dispersive regime but even in the resonant case. Though exact and computable, the expressions we get are too cumbersome to be displayed here and do not easily lend themselves to physical interpretation. In the next section, we introduce an effective non-linear three-boson model which allows us to derive the same results to the lowest order, and has also the advantage of being physically more transparent.

IV. EFFECTIVE NON-LINEAR THREE-BOSON MODEL

A. Non-linear absorption and dispersion in the quantum regime.

We consider a system of three bosons of respective annihilation operators a , b and c , whose non-linear Hamil-

tonian is given by

$$H = -\Delta_c a^\dagger a + \alpha (a + a^\dagger) - \Delta_e b^\dagger b - \Delta_r c^\dagger c + g\sqrt{N} (ab^\dagger + b^\dagger a) + \frac{\Omega_{cf}}{2} (bc^\dagger + b^\dagger c) + \frac{\kappa_r}{2} c^\dagger c^\dagger cc$$

We moreover assume that the c -boson is coupled to a *non-linear* bath whose action on the system is represented by the following non-linear dissipation operator, acting on the density matrix ρ of the system

$$\mathcal{D}[\rho] = \frac{\kappa_i}{2} \{2cc\rho c^\dagger c^\dagger - c^\dagger c^\dagger c\rho - \rho c^\dagger c^\dagger cc\}$$

Here, all parameters, in particular κ_r and κ_i , are assumed real. From the full Liouville-von Neumann equation of the system $\partial_t \rho = -\frac{i}{\hbar} [H, \rho] + \mathcal{D}[\rho]$ one readily derives the following Bloch equations

$$\begin{aligned} \frac{d}{dt} \langle a \rangle &= iD_c \langle a \rangle - i\alpha - ig\sqrt{N} \langle b \rangle \\ \frac{d}{dt} \langle b \rangle &= iD_e \langle b \rangle - ig\sqrt{N} \langle a \rangle - i\frac{\Omega_{cf}}{2} \langle c \rangle \\ \frac{d}{dt} \langle c \rangle &= iD_r \langle c \rangle - i\frac{\Omega_{cf}}{2} \langle b \rangle - i\kappa \langle c^\dagger cc \rangle \end{aligned}$$

where we introduced the notation $\kappa \equiv \kappa_r - i\kappa_i$. From this set of equations, one gets the same steady state value $\langle a \rangle^{(1)}$ as in Eq. (5). At the second order in α , the set of equations for two-operator steady-state averages is derived in the same way (here we omit superscripts ^(1,2) for simplicity)

$$\begin{aligned}
\langle aa \rangle &= \frac{g\sqrt{N}}{D_c} \langle ab \rangle + \frac{\alpha}{D_c} \langle a \rangle \\
\langle ab \rangle &= \frac{\Omega_{cf}}{2(D_c + D_e)} \langle ac \rangle + \frac{g\sqrt{N}}{(D_c + D_e)} \langle aa \rangle + \frac{g\sqrt{N}}{(D_c + D_e)} \langle bb \rangle + \frac{\alpha}{(D_c + D_e)} \langle b \rangle \\
\langle ac \rangle &= \frac{g\sqrt{N}}{(D_c + D_r)} \langle bc \rangle + \frac{\alpha}{(D_c + D_r)} \langle c \rangle + \frac{\Omega_{cf}}{2(D_c + D_r)} \langle ab \rangle \\
\langle bb \rangle &= \frac{\Omega_{cf}}{2D_e} \langle bc \rangle + \frac{g\sqrt{N}}{D_e} \langle ab \rangle \\
\langle bc \rangle &= \frac{\Omega_{cf}}{2(D_e + D_r)} \langle cc \rangle + \frac{g\sqrt{N}}{(D_e + D_r)} \langle ac \rangle + \frac{\Omega_{cf}}{2(D_e + D_r)} \langle bb \rangle \\
\langle cc \rangle &= \frac{\Omega_{cf}}{2(D_r - \frac{\kappa}{2})} \langle bc \rangle
\end{aligned}$$

which agrees with Eqs. (8-13) but for the last equation. If, however, we eliminate $\langle bc \rangle$ and $\langle bb \rangle$ from the last three equations, one obtains

$$\begin{aligned}
\langle cc \rangle &= \frac{1}{(D_r - \frac{\kappa}{2}) \left(D_r + D_e - \frac{\Omega_{cf}^2}{4D_e} \right) - \frac{\Omega_{cf}^2}{4}} \\
&\times \frac{\Omega_{cf}g\sqrt{N}}{2} \left(\langle ac \rangle + \frac{\Omega_{cf}}{2D_e} \langle ab \rangle \right)
\end{aligned}$$

which can be identified with Eq. (13) provided that

$$K = \frac{1}{(D_r - \frac{\kappa}{2}) \left(D_r + D_e - \frac{\Omega_{cf}^2}{4D_e} \right) - \frac{\Omega_{cf}^2}{4}}$$

which, upon recalling Eq. (14), yields

$$\kappa = 2 \left(\frac{V_b}{V - V_b} \right) \left(\frac{\Omega_{cf}^2}{4 \left(D_r + D_e - \frac{\Omega_{cf}^2}{4D_e} \right)} - D_r \right)$$

We obtain thus the analytic expressions of the parameters κ_r and κ_i , respectively characterizing the non-linear dispersion and absorption of the c -boson, which make our model system precisely reproduce the results of the original problem in the steady state and in the lowest order of the feeding parameter α .

B. Discussion.

Let us now investigate the physical content of the previous model by considering two limiting cases.

In the dispersive regime addressed in our previous work [6], $|D_{e,r}| \gg \Omega_{cf}$, whence $V_b \approx \frac{\sqrt{2}\pi^2}{3} \sqrt{\frac{|C_6|}{\Delta_r}}$, $\kappa_r \approx -\frac{2\Delta_r}{(N_b-1)}$ and $\kappa_i \approx 0$, where we introduced $N_b \equiv \frac{V}{V_b}$. This result agrees with what we previously obtained in

the Rydberg bubble approximation [6] and therefore confirms its validity: we observe a shift due to the non-linear dispersive behavior of the c -boson, but no non-linear absorption since the intermediate level is too far detuned. Moreover, in the bubble picture, the parameter N_b was interpreted as the number of Rydberg bubbles the sample may accommodate; as suggested above, this allows to interpret V_b as the bubble volume.

If we now go to the opposite regime, *i.e.* the resonant case for which $\Delta_e = \Delta_r = 0$, $\gamma_e \gg \gamma_r$ and $\Omega_{cf}^2 \gg \gamma_e^2$, we obtain $V_b \approx \frac{\pi^2}{3} (1 - i) \sqrt{\frac{|C_6|}{\gamma_e}}$ and therefore the non-linearity parameters are

$$\kappa_r = -\kappa_i \approx -\frac{2\pi^2}{3V} \sqrt{\gamma_e |C_6|}$$

We now have both dispersion *and* absorption. From the expression of κ_i , it is clear that absorption results from an interplay of the spontaneous emission from the intermediate state and the Rydberg-Rydberg interactions.

V. CONCLUSION

In this article, we have studied the strong quantum optical non-linearities induced by Rydberg-Rydberg van der Waals interactions in an atomic medium. We provided a new perturbative treatment of the problem, based on the factorization of the lowest perturbative order of operator product averages. Though being purely radiative damping, this factorization property is approximately preserved in the presence of to, e.g., laser frequency and intensity noise, as it is the case in our experimental setup. Our perturbative calculations enabled us to recover and extend our previous results: we could validate the approach based on the Rydberg bubble picture, as well as investigate the resonant, absorptive, regime. In particular, our numerical simulations showed that strong Rydberg-induced non-linearities led to different impedance matching conditions for single photons and photon pairs.

Moreover we proposed an effective model which leads to the same results as the full calculation at the lowest order in the feeding parameter; this model also sheds some light on the origin of the dispersion and absorption, as well as makes a bridge between the Rydberg bubble and perturbative approaches. In the future, we shall first try and take advantage of our understanding of the system to investigate regimes of parameters for which a photonic gate can be implemented. On the other hand, we also plan to apply other methods, inspired from many-body physics to the problem, in order to recover and further extend our results.

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Appendix A: The full Hamiltonian in the Rotating Wave Approximation

The full Hamiltonian of the system can be written under the form

$$\begin{aligned}
H &= H_{at} + H_{cav} + H_{bath} + V_{at-cav} + V_{cav-bath} + V_{at-bath} \\
H_{at} &\equiv \hbar\omega_e \sum_{n=1}^N \sigma_{ee}^{(n)} + \hbar\Omega_{cf} \cos(\omega_{cf}t) \sum_{n=1}^N \left(\sigma_{re}^{(n)} + \sigma_{er}^{(n)} \right) + \\
&\quad + \hbar\omega_r \sum_{n=1}^N \sigma_{rr}^{(n)} + \sum_{m < n=1}^N \hbar\kappa_{mn} \sigma_{rr}^{(m)} \sigma_{rr}^{(n)} \\
H_{cav} &\equiv \hbar\omega_c a^\dagger a \\
H_{bath} &\equiv \int d\omega \hbar\omega (b^\dagger b + c^\dagger c + d^\dagger d) \\
V_{at-cav} &\equiv \sum_{n=1}^N \hbar g (a + a^\dagger) \left(\sigma_{eg}^{(n)} + \sigma_{ge}^{(n)} \right) \\
V_{cav-bath} &\equiv \int d\omega \hbar g_b (b + b^\dagger) (a + a^\dagger) \\
V_{at-bath} &\equiv \int d\omega \hbar g_c (c + c^\dagger) \left(\sigma_{eg}^{(n)} + \sigma_{ge}^{(n)} \right) + \\
&\quad \int d\omega \hbar g_d (d + d^\dagger) \left(\sigma_{rg}^{(n)} + \sigma_{gr}^{(n)} \right)
\end{aligned}$$

where $\sigma_{\alpha\beta} \equiv |\alpha\rangle\langle\beta|$, $\hbar\omega_\alpha$ is the energy of the atomic level $|\alpha\rangle$ for $\alpha = e, r$ (with the convention $\omega_g = 0$), and $\kappa_{mn} \equiv C_6 / \|\vec{r}_m - \vec{r}_n\|^6$ denotes the van der Waals interaction between atoms in the Rydberg level – when atoms are in the ground or intermediate states, their interactions are neglected. The operators $b(\omega)$, $c(\omega)$ and $d(\omega)$ denoted simply as b , c , d , are bath operators coupled to the cavity and atomic operators with the respective coupling strengths $g_b(\omega)$, $g_c(\omega)$ and $g_d(\omega)$.

We switch to the rotating frame defined by $|\psi\rangle \rightarrow$

$$|\tilde{\psi}\rangle = \exp\left(-\frac{it}{\hbar} H_0\right) \text{ where}$$

$$\begin{aligned}
H_0 &\equiv \hbar\omega_p a^\dagger a + \sum_{n=1}^N \left(\hbar\omega_p \sigma_{ee}^{(n)} + \hbar(\omega_p + \omega_{cf}) \sigma_{rr}^{(n)} \right) + \\
&\quad \int d\omega \left(\hbar\omega_p b^\dagger b + \hbar(\omega_p + \omega_{cf}) c^\dagger c + \hbar\omega_p d^\dagger d \right)
\end{aligned}$$

and perform the Rotating Wave Approximation to get the new Hamiltonian

$$\begin{aligned}
\tilde{H} &= \tilde{H}_{at} + \tilde{H}_{cav} + \tilde{H}_{bath} + \tilde{V}_{at-cav} + \tilde{V}_{at-bath} + \tilde{V}_{cav-bath} \\
\tilde{H}_{at} &= -\hbar\Delta_e \sum_{n=1}^N \sigma_{ee}^{(n)} + \sum_{m < n=1}^N \hbar\kappa_{mn} \sigma_{rr}^{(m)} \sigma_{rr}^{(n)} + \\
&\quad -\hbar\Delta_r \sum_{n=1}^N \sigma_{rr}^{(n)} + \frac{\hbar\Omega_{cf}}{2} \sum_{n=1}^N \left(\sigma_{re}^{(n)} + \sigma_{er}^{(n)} \right) \\
\tilde{H}_{cav} &= -\hbar\Delta_c a^\dagger a \\
\tilde{H}_{bath} &\approx \int d\omega \hbar\omega (b(\omega + \omega_p))^\dagger b(\omega + \omega_p) + \\
&\quad \int d\omega \hbar\omega \sum_{n=1}^N (c_n(\omega + \omega_p + \omega_{cf}))^\dagger c_n(\omega + \omega_p + \omega_{cf}) + \\
&\quad \int d\omega \hbar\omega \sum_{n=1}^N (d_n(\omega + \omega_p))^\dagger d_n(\omega + \omega_p) \\
\tilde{V}_{a-c} &\approx \sum_{n=1}^N \hbar g \left(a \sigma_{eg}^{(n)} + a^\dagger \sigma_{ge}^{(n)} \right) \\
\tilde{V}_{cav-bath} &\approx \int d\omega \hbar g_b(\omega) \left[b(\omega) a^\dagger + (b(\omega))^\dagger a \right] \\
\tilde{V}_{at-bath} &\approx \sum_{n=1}^N \int d\omega \hbar g_c(\omega) \left[c_n(\omega) \sigma_{eg}^{(n)} + (c_n(\omega))^\dagger \sigma_{ge}^{(n)} \right] + \\
&\quad \sum_{n=1}^N \int d\omega \hbar g_d(\omega) \left[d_n(\omega) \sigma_{rg}^{(n)} + (d_n(\omega))^\dagger \sigma_{gr}^{(n)} \right]
\end{aligned}$$

with the detunings $\Delta_c \equiv (\omega_p - \omega_c)$, $\Delta_e \equiv (\omega_p - \omega_e)$, and $\Delta_r \equiv (\omega_p + \omega_{cf} - \omega_r)$. It is important to note that the evolution under the Hamiltonian \tilde{H} conserves the number of excitations.

Appendix B: Factorization of correlation functions.

We suppose that the bath interacting with the cavity is initially in the following continuous-mode coherent state (incoming quasi-classical field)

$$|\alpha\rangle = e^{-\frac{1}{2}\langle n \rangle} e^{\sqrt{\langle n \rangle} b_\alpha^\dagger} |0\rangle$$

where $\int |\alpha(t)|^2 dt = \langle n \rangle$ and $b_\alpha^\dagger = \frac{1}{\sqrt{\langle n \rangle}} \int d\omega \alpha(\omega) b^\dagger(\omega)$ is a superposition of bath mode creation operators $b^\dagger(\omega)$ [14]. Note that with this definition, b_α is a bosonic operator, *i.e.* $[b_\alpha, b_\alpha^\dagger] = 1$. The atoms and cavity field are initially in their ground state denoted by $|G\rangle \equiv |g \dots g\rangle \otimes |0\rangle$.

Let us consider, for instance, the quantity $\langle \alpha, G | a^\dagger(t_1) a^\dagger(t_2) a(t_2) a(t_1) | G, \alpha \rangle$, for $t_2 > t_1$, where $|G, \alpha\rangle$ denotes the initial state of the whole system {atoms+cavity+baths}, the baths coupled to the atoms are supposed empty and their state is not explicitly written,

$$\begin{aligned} & \langle \alpha, G | a^\dagger(t_1) a^\dagger(t_2) a(t_2) a(t_1) | G, \alpha \rangle \\ &= e^{-\langle n \rangle} \sum_{k,l} \frac{\langle n \rangle^{\frac{k+l}{2}}}{\sqrt{k!l!}} \langle k, G | a^\dagger(t_1) a^\dagger(t_2) a(t_2) a(t_1) | G, l \rangle \end{aligned} \quad (\text{B1})$$

Expanding this expression with respect to $|\alpha|$ (which is equivalent to expanding in the number of excitations present in the system), one finds that the lowest non-vanishing contribution is the fourth order term $k = l = 2$. For the system considered the identity operator can be represented in the following way $\mathcal{I} = \bigotimes_i \mathcal{I}_i$ where $\mathcal{I}_i = \sum_q |q_i\rangle \langle q_i|$ are the identity operators on each degree of freedom of the system, and $|q_i\rangle$'s denote q -th basis vector of i -th degree of freedom. Inserting this identity operator between $a^\dagger(t_2)$ and $a(t_2)$ of the quantity (B1) yields:

$$\begin{aligned} & e^{-\langle n \rangle} \sum_{k,l} \frac{\langle n \rangle^{\frac{k+l}{2}}}{\sqrt{k!l!}} \langle k, G | a^\dagger(t_1) a^\dagger(t_2) \\ & \left\{ \bigotimes_i \sum_q |q_i\rangle \langle q_i| \right\} a(t_2) a(t_1) | G, l \rangle \end{aligned} \quad (\text{B2})$$

For the lowest non-vanishing term $k = 2, l = 2$:

$$a(t_2) a(t_1) | G, 2 \rangle = e^{i\frac{\tilde{H}t_2}{\hbar}} a e^{i\frac{\tilde{H}}{\hbar}(t_1-t_2)} a | G, 2(t_1) \rangle$$

where $|G, 2(t_1)\rangle \equiv e^{-i\frac{\tilde{H}t_1}{\hbar}} |G, 2\rangle$ (note that this state can contain excited atoms and/or cavity photons). The state $a | G, 2(t_1) \rangle$ can at most contain one excitation, and so can the state $e^{iH'(t_1-t_2)} a | G, 2(t_1) \rangle$ due to the conservation of excitation number. Hence $e^{iH't_2} a e^{iH'(t_1-t_2)} a | G, 2(t_1) \rangle$ can only have component on $|G, 0\rangle$. Finally the fourth order expression of (B2) reads:

$$\begin{aligned} & e^{-\langle n \rangle} \frac{\langle n \rangle^2}{2} \langle 2, G | a^\dagger(t_1) a^\dagger(t_2) a(t_2) a(t_1) | G, 2 \rangle \\ &= e^{-\langle n \rangle} \frac{\langle n \rangle^2}{2} |\langle 2, G | a^\dagger(t_1) a^\dagger(t_2) | G, 0 \rangle|^2 \\ &= \langle \alpha, G | a^\dagger(t_1) a^\dagger(t_2) | G, 0 \rangle_2 \langle G, 0 | a(t_2) a(t_1) | G, \alpha \rangle_2 \end{aligned}$$

where we used that $e^{-\frac{\langle n \rangle}{2}} \frac{\langle n \rangle}{\sqrt{2}} \langle 2, G | a^\dagger(t_1) a^\dagger(t_2) | G, 0 \rangle$ and $e^{-\frac{\langle n \rangle}{2}} \frac{\langle n \rangle}{\sqrt{2}} \langle 0, G | a(t_2) a(t_1) | G, 2 \rangle$ are equal to the second order expansion in $|\alpha|$ of quantities $\langle \alpha, G | a^\dagger(t_1) a^\dagger(t_2) | G, \alpha \rangle$ and $\langle \alpha, G | a^\dagger(t_1) a^\dagger(t_2) | G, \alpha \rangle$ respectively, which we denoted by $\langle \dots \rangle_2$.

Thus to compute $\langle \alpha, G | a^\dagger(t_1) a^\dagger(t_2) a(t_2) a(t_1) | G, \alpha \rangle$ in the lowest order it is enough to calculate $\langle a(t_2) a(t_1) \rangle \equiv \langle \alpha, G | a(t_2) a(t_1) | G, \alpha \rangle$.

The same argument holds for more general mean values such as

$$\begin{aligned} & \langle \alpha, G | a^\dagger(t_1) a^\dagger(t_2) \dots a^\dagger(t_p) a(t_{p+1}) \dots \\ & \dots a(t_{p+q-1}) a(t_{p+q}) | G, \alpha \rangle^{(p+q)} \end{aligned}$$

and in particular

$$\begin{aligned} & \langle a^\dagger(t) a(t) \rangle^{(2)} = \langle a^\dagger(t) \rangle^{(1)} \langle a(t) \rangle^{(1)} \\ & \langle a^\dagger(t_2) a^\dagger(t_1) a(t_1) \rangle^{(3)} = \langle a^\dagger(t_2) a^\dagger(t_1) \rangle^{(2)} \\ & \quad \times \langle a(t_1) \rangle^{(1)} \\ & \langle a^\dagger(t_2) a^\dagger(t_1) a(t_1) a(t_2) \rangle^{(4)} = \langle a^\dagger(t_2) a^\dagger(t_1) \rangle^{(2)} \\ & \quad \times \langle a(t_1) a(t_2) \rangle^{(2)} \end{aligned}$$

Appendix C: Calculation of $\langle aa \rangle^{(2)}$

The system of equations for the same-time 2-operator products in the second order in α is readily derived from Heisenberg-Langevin equations. For notational convenience here, we do not explicitly write superscripts $^{(1,2)}$, nor the time since we only dealt with same-time mean values : hence $\langle aa \rangle$ should be understood as $\langle a(t) a(t) \rangle^{(2)}$ and $\langle \sigma_{ge}^{(i)} \rangle$ as $\langle \sigma_{ge}^{(i)}(t) \rangle^{(1)}$. We thus find

$$\begin{aligned}
\frac{d}{dt} \langle aa \rangle &= 2D_c \langle aa \rangle - 2ig \sum_i \langle a\sigma_{ge}^{(i)} \rangle - 2i\alpha \langle a \rangle \\
\frac{d}{dt} \langle a\sigma_{ge}^{(i)} \rangle &= (D_c + D_e) \langle a\sigma_{ge}^{(i)} \rangle - i\frac{\Omega_b}{2} \langle a\sigma_{gr}^{(i)} \rangle - ig \langle aa \rangle - ig \sum_j \langle \sigma_{ge}^{(j)} \sigma_{ge}^{(i)} \rangle - i\alpha \langle \sigma_{ge}^{(i)} \rangle \\
\frac{d}{dt} \langle a\sigma_{gr}^{(i)} \rangle &= (D_c + D_r) \langle a\sigma_{gr}^{(i)} \rangle - ig \sum_j \langle \sigma_{ge}^{(j)} \sigma_{gr}^{(i)} \rangle - i\alpha \langle \sigma_{gr}^{(i)} \rangle - i\frac{\Omega_b}{2} \langle a\sigma_{ge}^{(i)} \rangle \\
\frac{d}{dt} \langle \sigma_{ge}^{(j)} \sigma_{ge}^{(i)} \rangle &= 2D_e \langle \sigma_{ge}^{(j)} \sigma_{ge}^{(i)} \rangle - i\frac{\Omega_b}{2} \langle \sigma_{ge}^{(j)} \sigma_{gr}^{(i)} \rangle - i\frac{\Omega_b}{2} \langle \sigma_{gr}^{(j)} \sigma_{ge}^{(i)} \rangle - ig \langle a\sigma_{ge}^{(j)} \rangle - ig \langle a\sigma_{ge}^{(i)} \rangle \\
\frac{d}{dt} \langle \sigma_{ge}^{(j)} \sigma_{gr}^{(i)} \rangle &= (D_e + D_r) \langle \sigma_{ge}^{(j)} \sigma_{gr}^{(i)} \rangle - i\frac{\Omega_b}{2} \langle \sigma_{gr}^{(j)} \sigma_{gr}^{(i)} \rangle - ig \langle a\sigma_{gr}^{(j)} \rangle - i\frac{\Omega_b}{2} \langle \sigma_{ge}^{(j)} \sigma_{ge}^{(i)} \rangle \\
\frac{d}{dt} \langle \sigma_{gr}^{(j)} \sigma_{gr}^{(i)} \rangle &= (2D_r - i\kappa_{i,j}) \langle \sigma_{gr}^{(j)} \sigma_{gr}^{(i)} \rangle - i\frac{\Omega_b}{2} \langle \sigma_{ge}^{(j)} \sigma_{gr}^{(i)} \rangle - i\frac{\Omega_b}{2} \langle \sigma_{gr}^{(j)} \sigma_{ge}^{(i)} \rangle
\end{aligned}$$

Assuming that the medium is homogeneous, *i.e.* that for all (i, j) , $\langle \sigma_{ge}^{(j)} \sigma_{gr}^{(i)} \rangle = \langle \sigma_{ge}^{(i)} \sigma_{gr}^{(j)} \rangle$ and $\langle a\sigma_{ge}^{(i)} \rangle =$

$\langle a\sigma_{ge}^{(j)} \rangle$, in the steady state this system yields

$$\begin{aligned}
\langle aa \rangle &= \frac{g}{D_c} \sum_i \langle a\sigma_{ge}^{(i)} \rangle + \frac{\alpha}{D_c} \langle a \rangle \\
\langle a\sigma_{ge}^{(i)} \rangle &= \frac{\Omega_b}{2(D_c + D_e)} \langle a\sigma_{gr}^{(i)} \rangle + \frac{g}{(D_c + D_e)} \langle aa \rangle + \frac{g}{(D_c + D_e)} \sum_j \langle \sigma_{ge}^{(j)} \sigma_{ge}^{(i)} \rangle + \frac{\alpha}{(D_c + D_e)} \langle \sigma_{ge}^{(i)} \rangle \\
\langle a\sigma_{gr}^{(i)} \rangle &= \frac{g}{(D_c + D_r)} \sum_j \langle \sigma_{ge}^{(j)} \sigma_{gr}^{(i)} \rangle + \frac{\alpha}{(D_c + D_r)} \langle \sigma_{gr}^{(i)} \rangle + \frac{\Omega_b}{2(D_c + D_r)} \langle a\sigma_{ge}^{(i)} \rangle \\
\langle \sigma_{ge}^{(j)} \sigma_{ge}^{(i)} \rangle &= \frac{\Omega_b}{2D_e} \langle \sigma_{ge}^{(j)} \sigma_{gr}^{(i)} \rangle + \frac{g}{D_e} \langle a\sigma_{ge}^{(i)} \rangle \\
\langle \sigma_{ge}^{(j)} \sigma_{gr}^{(i)} \rangle &= \frac{\Omega_b}{2(D_e + D_r)} \langle \sigma_{gr}^{(j)} \sigma_{gr}^{(i)} \rangle + \frac{g}{(D_e + D_r)} \langle a\sigma_{gr}^{(i)} \rangle + \frac{\Omega_b}{2(D_e + D_r)} \langle \sigma_{ge}^{(j)} \sigma_{ge}^{(i)} \rangle \\
\langle \sigma_{gr}^{(j)} \sigma_{gr}^{(i)} \rangle &= \frac{\Omega_b}{2(D_r - \frac{\kappa_{i,j}}{2})} \langle \sigma_{ge}^{(j)} \sigma_{gr}^{(i)} \rangle
\end{aligned}$$

Note that the first-order values $\langle a \rangle \equiv \langle a \rangle^{(1)}$, $\langle \sigma_{ge}^{(i)} \rangle \equiv \langle \sigma_{ge}^{(i)} \rangle^{(1)}$, $\langle \sigma_{gr}^{(i)} \rangle \equiv \langle \sigma_{gr}^{(i)} \rangle^{(1)}$ have been determined through solving the first-order steady state system, see Eqs. (5-7) in the main text.

Summing the above equations over atom numbers (i, j) yields a system on averages of the collective operators $b \equiv \frac{1}{\sqrt{N}} \sum_i \sigma_{ge}^{(i)}$ and $c \equiv \frac{1}{\sqrt{N}} \sum_i \sigma_{gr}^{(i)}$ and field operator a , which is *almost* closed but for the last equation which will now be considered and approximated. Eliminating $\langle \sigma_{ge}^{(j)} \sigma_{gr}^{(i)} \rangle$ and $\langle \sigma_{ge}^{(j)} \sigma_{ge}^{(i)} \rangle$ from the last three equations

we get

$$\begin{aligned}
\langle \sigma_{gr}^{(j)} \sigma_{gr}^{(i)} \rangle &= \\
&= \frac{\Omega_b g}{2 \left\{ \left(D_r - \frac{\kappa_{i,j}}{2} \right) \left[(D_e + D_r) - \frac{\Omega_b^2}{4D_e} \right] - \frac{\Omega_b^2}{4} \right\}} \langle a\sigma_{gr}^{(i)} \rangle \\
&+ \frac{\Omega_b^2 g}{4D_e \left\{ \left(D_r - \frac{\kappa_{i,j}}{2} \right) \left[(D_e + D_r) - \frac{\Omega_b^2}{4D_e} \right] - \frac{\Omega_b^2}{4} \right\}} \langle a\sigma_{ge}^{(i)} \rangle
\end{aligned}$$

We now sum over i and j indices and divide by N this equation to get

$$\langle cc \rangle = \frac{\Omega_b g}{2} \sum_i K_i \langle a\sigma_{gr}^{(i)} \rangle + \frac{\Omega_b^2 g}{4D_e} \sum_i K_i \langle a\sigma_{ge}^{(i)} \rangle$$

where we introduced the coefficient

$$K_i \equiv \frac{1}{N} \sum_j \frac{1}{\left(D_e + D_r - \frac{\Omega_b^2}{4D_e}\right) \left(D_r - \frac{\kappa_{i,j}}{2}\right) - \frac{\Omega_b^2}{4}}.$$

Making the approximation that K_i does not depend on i , *i.e.* $K_i \approx K$, we get:

$$\langle cc \rangle \approx \frac{\Omega_b g \sqrt{N}}{2} K \langle ac \rangle + \frac{\Omega_b^2 g \sqrt{N}}{4D_e} K \langle ab \rangle$$

To estimate K we consider that the sample is a sphere of radius R

$$\begin{aligned} K &= \frac{1}{N} \sum_j \frac{1}{\left(D_e + D_r - \frac{\Omega_b^2}{4D_e}\right) \left(D_r - \frac{\kappa_{i,j}}{2}\right) - \frac{\Omega_b^2}{4}} \\ &\approx \frac{4\pi}{\frac{4}{3}R^3} \int_0^R \frac{r^2}{\left(D_e + D_r - \frac{\Omega_b^2}{4D_e}\right) \left(D_r - \frac{C_6}{2r^6}\right) - \frac{\Omega_b^2}{4}} dr \\ &= \frac{3}{R^3} \int_0^R \frac{r^2}{\left(D_e + D_r - \frac{\Omega_b^2}{4D_e}\right) \left(D_r - \frac{C_6}{2r^6}\right) - \frac{\Omega_b^2}{4}} dr \end{aligned}$$

For large values of R , K does not depend on the geometry

$$\begin{aligned} K &\underset{R \rightarrow \infty}{\sim} \frac{1}{\left(D_e + D_r - \frac{\Omega_b^2}{4D_e}\right) D_r - \frac{\Omega_b^2}{4}} \\ &\times \left(1 - \frac{\sqrt{2}\pi^2}{3V} \sqrt{\frac{C_6}{\frac{\Omega_b^2}{4\left(D_e + D_r - \frac{\Omega_b^2}{4D_e}\right)} - D_r}} \right) \end{aligned}$$

Finally the desired closed system is

$$\begin{aligned} \langle aa \rangle &= \frac{g\sqrt{N}}{D_c} \langle ab \rangle + \frac{\alpha}{D_c} \langle a \rangle \\ \langle ab \rangle &= \frac{\Omega_b}{2(D_c + D_e)} \langle ac \rangle + \frac{g\sqrt{N}}{(D_c + D_e)} \langle aa \rangle + \frac{g\sqrt{N}}{(D_c + D_e)} \langle bb \rangle + \frac{\alpha}{(D_c + D_e)} \langle b \rangle \\ \langle ac \rangle &= \frac{g\sqrt{N}}{(D_c + D_r)} \langle bc \rangle + \frac{\alpha}{(D_c + D_r)} \langle c \rangle + \frac{\Omega_b}{2(D_c + D_r)} \langle ab \rangle \\ \langle bb \rangle &= \frac{\Omega_b}{2D_e} \langle bc \rangle + \frac{g\sqrt{N}}{D_e} \langle ab \rangle \\ \langle bc \rangle &= \frac{\Omega_b}{2(D_e + D_r)} \langle cc \rangle + \frac{g\sqrt{N}}{(D_e + D_r)} \langle ac \rangle + \frac{\Omega_b}{2(D_e + D_r)} \langle bb \rangle \\ \langle cc \rangle &= \frac{\Omega_b g \sqrt{N}}{2} K \langle ac \rangle + \frac{\Omega_b^2 g \sqrt{N}}{4D_e} K \langle ab \rangle \end{aligned} \tag{C1}$$

which allows to determine $\langle aa \rangle$. The analytical solution is too cumbersome to be displayed in this paper but can be readily obtained by matrix inversion.

Appendix D: Factorization in the presence of extra dephasing

In this appendix, we show in which conditions the factorization of field operator products described in Appendix B remains valid in the presence of extra dephasing due to laser frequency and intensity noise. Such dephasing is correctly accounted for by adding the term $-\gamma_d \sigma_{gr}^{(n)} + F_{gr}^{(d)}$ in the Heisenberg-Langevin equation Eq. (3) on $\sigma_{gr}^{(n)}$, where $F_{gr}^{(d)}$ is an extra Langevin force and

$\gamma_d \approx 0.15 \times \gamma_e$, $\gamma_r \approx 0.01 \times \gamma_e$ and $\gamma_e = 2\pi \times 3$ MHz in the experimental setup.

In the absence of interatomic interactions, because laser and cavity fields address the atoms symmetrically, the ensemble evolves in the subspace of symmetric states. The atomic system essentially remains in this subspace, even when the interactions are taken into account, if the number of Rydberg excitations in the sample is much less than the total number of Rydberg bubbles the ensemble can accomodate for. Such symmetric superpositions actually not only contain “allowed” components (*i.e.* with Rydberg atoms further than a Rydberg bubble radius apart) but also “forbidden” components (with Rydberg atoms closer than a Rydberg bubble radius). Their number is, however, very small compared to that of “allowed”

configurations and they will therefore only slightly alter the outcome of dissipative dynamics of the system.

Under these assumptions, let us show in which conditions the mean value $\langle c^\dagger c \rangle$ factorizes at lowest order. Focusing on the dissipative part of Bloch equations for $\sigma_{gr}^{(i)}$ and $\sigma_{rr}^{(i)}$ (note that for the latter, there is no extra dephasing) we get

$$\begin{aligned}\frac{d}{dt} \langle \sigma_{gr}^{(i)} \rangle \Big|_d &= -(\gamma_r + \gamma_d) \langle \sigma_{gr}^{(i)} \rangle \\ \frac{d}{dt} \langle \sigma_{rg}^{(i)} \sigma_{gr}^{(j)} \rangle \Big|_{d, i \neq j} &= -2(\gamma_r + \gamma_d) \langle \sigma_{rg}^{(i)} \sigma_{gr}^{(j)} \rangle \\ \frac{d}{dt} \langle \sigma_{rr}^{(i)} \rangle \Big|_d &= -2\gamma_r \langle \sigma_{rr}^{(i)} \rangle\end{aligned}$$

and recalling that $c \equiv \frac{1}{\sqrt{N}} \sum_i \sigma_{gr}^{(i)}$, we get $\langle c^\dagger c \rangle = \frac{1}{N} \sum_i \langle \sigma_{rr}^{(i)} \rangle + \frac{1}{N} \sum_{i \neq j} \langle \sigma_{rg}^{(i)} \sigma_{gr}^{(j)} \rangle$ whence, for a short time interval

$$\begin{aligned}\frac{d \langle c^\dagger c \rangle}{dt} \Big|_d &= \frac{1}{N} \sum_i \frac{d}{dt} \langle \sigma_{rr}^{(i)} \rangle \Big|_d + \frac{1}{N} \sum_{i \neq j} \frac{d}{dt} \langle \sigma_{rg}^{(i)} \sigma_{gr}^{(j)} \rangle \Big|_d \\ &= -\frac{2\gamma_r}{N} \sum_i \langle \sigma_{rr}^{(i)} \rangle - \frac{2}{N} (\gamma_r + \gamma_d) \sum_{i \neq j} \langle \sigma_{rg}^{(i)} \sigma_{gr}^{(j)} \rangle \\ &= -\frac{2\gamma_r}{N} \sum_i \langle \sigma_{rr}^{(i)} \rangle + \frac{2}{N} (\gamma_r + \gamma_d) \sum_i \langle \sigma_{rr}^{(i)} \rangle + \\ &\quad - \frac{2}{N} (\gamma_r + \gamma_d) \sum_{i,j} \langle \sigma_{rg}^{(i)} \sigma_{gr}^{(j)} \rangle \\ &= \frac{2\gamma_d}{N} \sum_i \langle \sigma_{rr}^{(i)} \rangle - \frac{2}{N} (\gamma_r + \gamma_d) \sum_{i,j} \langle \sigma_{rg}^{(i)} \sigma_{gr}^{(j)} \rangle \\ \frac{d}{dt} \langle c^\dagger c \rangle \Big|_d &= \frac{2\gamma_d}{N} \sum_i \langle \sigma_{rr}^{(i)} \rangle - 2(\gamma_r + \gamma_d) \langle c^\dagger c \rangle\end{aligned}$$

When there are n_r Rydberg excitations in the sample, with $n_r \ll N_b \ll N$ (N_b is the maximum number of Rydberg excitations the sample can contain), one has $\langle c^\dagger c \rangle \approx \sum_i \langle \sigma_{rr}^{(i)} \rangle \approx n_r$ whence

$$\frac{d}{dt} \langle c^\dagger c \rangle \Big|_d \approx -2 \left[\gamma_r + \gamma_d \left(1 - \frac{1}{N} \right) \right] \langle c^\dagger c \rangle$$

and for $\gamma_r \ll \gamma_d \ll N\gamma_r$

$$\frac{d}{dt} \langle c^\dagger c \rangle \Big|_d \approx -2\gamma_d \langle c^\dagger c \rangle$$

so, from the point of view of $c^\dagger c$, everything works as if the system was radiatively damped with the rate γ_d . In the same conditions, we moreover have

$$\frac{d}{dt} \langle c \rangle \Big|_d \approx -\gamma_d \langle c \rangle$$

and again, from the point of view of c , everything works as if the system was radiatively damped with the rate γ_d . Moreover, since all other dynamical equations (for population, coherence and field operator mean values) remain formally the same as in the purely radiative damping, the factorization procedure remains valid for $\langle a^\dagger a \rangle$ provided that $\gamma_r \ll \gamma_d \ll N\gamma_r$ and the radiative coherence decay γ_r is effectively replaced by the dephasing decay rate γ_d .

This result can also be extended to higher order quantities $\langle (a^\dagger)^m a^p \rangle$.

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